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Research Studies on Extreme Ultraviolet and Soft X-Ray Lasers

Annual Technical Report for the Period

1 September 1986 — 31 August 1987



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Section 1

Introduction

The goal of this program is to investigate, both theoretically and experimentally, new approaches to constructing XUV and soft x-ray lasers and also to develop new spectroscopic techniques for the study of this spectral region. This last work period has been one of great success for our efforts in both of these areas.

The most exciting of the recent results is the development of a fully saturated 109 nm photoionization pumped laser. Using only 3.5 J in a 300 ps, 1064 nm pump pulse, we measure a total small signal gain of $\exp(40)$. The laser uses a unique traveling wave geometry, which employs a blazed target. This allows near synchronism between an effective traveling wave plasma and the generated 109 nm beam. At present, this laser produces an output energy of $20 \,\mu\text{J}$ in a beam with a pulse-width of about 50 ps and with a divergence of 10 mrad. The paper describing this work is included as appendix 1 to this report.

Another key result during this period was the observation of large gains [exp.(5.1)] in a photoionization pumped super Coster Kronig system in Zn III. This super Coster Kronig type system, proposed earlier by Mendelsohn and Harris under this contract, is characterized by especially high selectivity. About 27% of the absorbed, broad-band, soft x-ray pumping radiation is channeled into the upper laser level in Zn III.

It should be stressed that the above systems are the first anywhere which show such large gains when pumped by small, and therefore, potentially high repetition rate laser systems. The 109 nm laser has the highest output power of any laser running in this spectral region.

Our spectroscopy project, where we use a combination of soft x-ray pumping and laser depletion to identify and measure core-excited autoionizing levels, also continues to go well. Following the recent Letter (J. K. Spong, J. D. Kmetec, S. C. Wallace, J. F. Young,

and S. E. Harris, "Laser Spectroscopy of Core-Excited Levels of Neutral Rubidium" *Phys. Rev. Lett.* 58, 2631–2634 (June 1987) we have continued to develop this technique so as to allow the measurement of autoionizing linewidths which lie beneath the Doppler widths of accessed levels. We have now extended this technique to allow a method of measurement that is independent of both the hyperfine structure and also of the Doppler width of the accessed species.

Section 2 of this report will summarize the cumulative contributions of this contract. Section 3 lists publications which have originated during this period.

We note that portions of this program have been jointly supported by the Office of Naval Research, Lawrence Livermore Laboratory, and the Strategic Defense Initiative Organization.

Section 2

Summary of Accomplishments

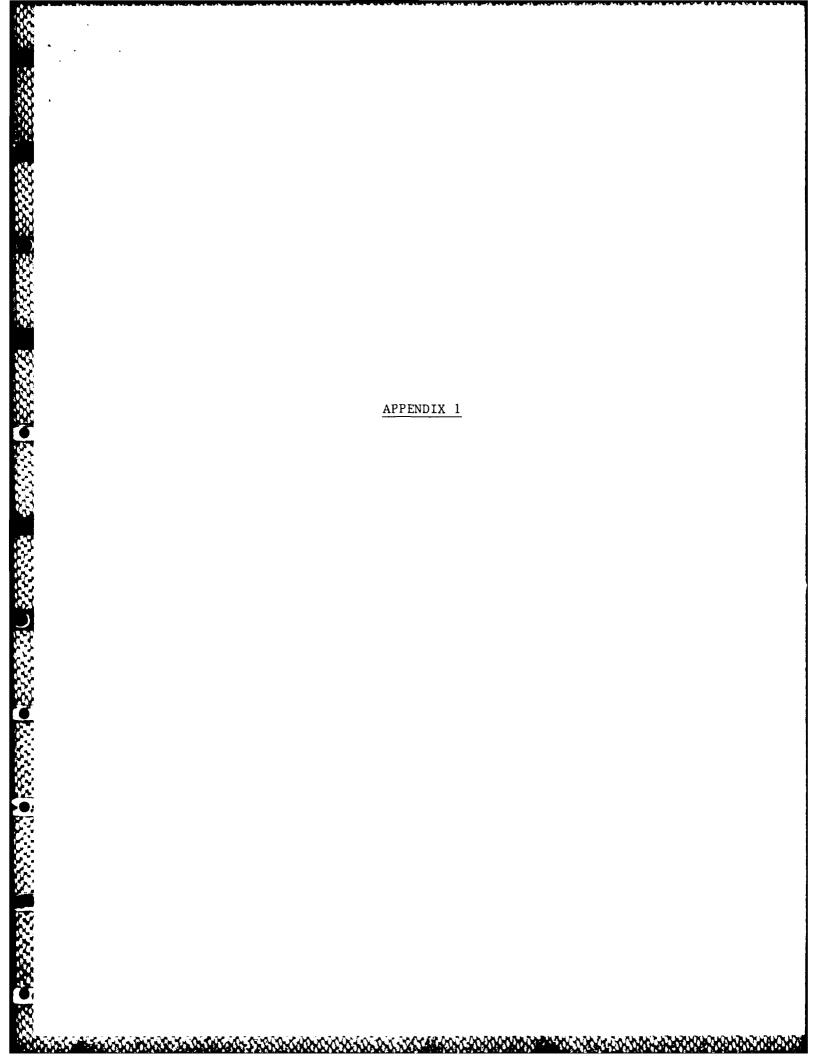
The summary of the accomplishments on this program during this (1 year) contract period follow:

- 1. We have completed and published our work on quasi-metastability. This concept, wherein we can identify radiating levels will be of importance, not only in the XUV, but also in the soft x-ray and harder x-ray spectral region.
- 2. A review paper "Core-Excited Metastable Levels: Application to Spectroscopy, to the Generation of Picosecond Extreme-Ultraviolet Pulses, and to Lasers" (S. E. Harris, and J. F. Young) was written and published.
- 3. As noted in the introduction, a new spectroscopic technique for study of core-excited levels was extended.
- 4. Also as noted in the introduction, we have studied the properties of the 109 nm Xe Auger laser and have used a new geometry to allow fully saturated operation of this device.
- 5. We have demonstrated high gain in the Zn III Auger laser.
- 6. A theoretical paper, where we use a new approach to study strong field laser induced ionization, was completed.

Section 3

Publications

- 1. D. P. Dimiduk, "Satellite Two-electron Radiative Transitions due to Relaxation Effects", Phys. Rev. A 35, 2338-2341 (March 1987).
- A. J. Mendelsohn, C. P. J. Barty, M. H. Sher, J. F. Young, and S. E. Harris, "Emission Spectra of Quasi Metastable Levels of Alkali-Metal Atoms," *Phys. Rev. A* 35, 2095– 2101 (March 1987).
- 3. S. E. Harris, and J. F. Young, "Core-Excited Metastable Levels: Application to Spectroscopy, to the Generation of Picosecond XUV Pulses, and to Lasers," J. Opt. Soc. B 4, 547-562 (April 1987).
- J. K. Spong, J. D. Kmetec, S. C. Wallace, J. F. Young, and S. E. Harris, "Laser Spectroscopy of Core-Excited Levels of Neutral Rubidium," *Phys. Rev. Lett.* 58, 2631-2634 (June 1987).
- 5. Guang-Yu Yin, C. P. J. Barty, D. A. King, D. J. Walker, S. E. Harris, and J. F. Young, "Low Energy Pumping of a 108.9 nm Xe Auger Laser," Opt. Lett. 12, 331-333 (May 1987).
- D. A. King, and R. G. Caro, "A Fast, High-Current Pulsed Discharge Device for the Inner-Shell Excitation of Atoms and Ions," IEEE J. Quant. Elect. QE-23, 418-425 (April 1987).
- 7. S. E. Harris, and J. K. Spong, "Laser Depletion Spectroscopy of Core-Excited Levels," in Laser Spectroscopy VIII (to be published).
- 8. M. H. Sher, J. J. Macklin, J. F. Young, and S. E. Harris, "Saturation of the Xe III 109 nm Laser Using Traveling-Wave Laser-Produced-Plasma Excitation," Opt. Lett. (to be published).
- 9. D. J. Walker, C. P. J. Barty, G. Y. Yin, J. F. Young, and S. E. Harris, "Observation of Super Coster-Kronig Pumped Gain in Zn III," Opt. Lett. (to be published).
- 10. R. Buffa, "An Approach to the Study of Strong-Field Laser-Induced Autoionization" (submitted for publication).



Saturation of the Xe III 109 nm Laser Using Traveling-Wave Laser-Produced-Plasma Excitation*

M. H. Sher, J. J. Macklin, J. F. Young, and S. E. Harris

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Abstract

We describe the construction and operation of a 109 nm, photoionization-pumped, single-pass laser in Xe III. The laser is pumped by soft x-rays emitted from a laser-produced plasma in a traveling-wave geometry. Using a 3.5 J, 300 psec, 1064 nm laser pump pulse, we measure a small-signal gain coefficient of $4.4\,\mathrm{cm^{-1}}$ and a total small signal gain of $\exp(40)$. The laser is fully saturated and produces an output energy of $20\,\mu\mathrm{J}$ in a beam with 10 mrad divergence.

^{*} This work was jointly supported by the U.S. Office of Naval Research, the U.S. Air Force Office of Scientific Research, the U.S. Army Research Office, the Strategic Defense Initiative Organization, and Lawrence Livermore National Laboratories.

Saturation of the Xe III 109 nm Laser Using Traveling-Wave Laser-Produced-Plasma Excitation

M. H. Sher, J. J. Macklin, J. F. Young, and S. E. Harris

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This Letter describes the construction and operation of a single-pass, $109 \,\mathrm{nm}$, Xe III Auger laser. The laser is pumped by soft x-rays, which are emitted from a laser-produced plasma in a traveling-wave geometry. Using only 3.5 J of $1064 \,\mathrm{nm}$ pump energy in a 300 psec pulse, we measure a small-signal gain coefficient of $4.4 \,\mathrm{cm}^{-1}$ and a total small-signal gain of $\exp(40)$. The $109 \,\mathrm{nm}$ laser is fully saturated over the second half of its length and produces an output energy of $20 \,\mu\mathrm{J}$ in a beam with $10 \,\mathrm{mrad}$ divergence.

Population inversion of the 109 nm transition was proposed and demonstrated by Kapteyn et al. ^{1,2} The inversion mechanism, outlined in the energy level diagram of Fig. 1, is inner-shell photoionization of a 4d electron, followed by Auger decay to Xe III. In this system, the Auger branching ratio is about 5% to both the upper and lower laser levels. The inversion results from the higher degeneracy of the lower level. Assuming only Doppler broadening, and ignoring hyperfine splitting, the gain cross section is 3×10^{-13} cm².

Proposals for photoionization pumping of short wavelength lasers and for Auger-pumped short wavelength lasers were made by Duguay³ and by McGuire.⁴ The possibility of constructing such lasers at low pumping energies was delineated by the work of Caro et al.,⁵ Silfvast et al.,⁶ and Mendelsohn and Harris.⁷ Recently, Yin et al.⁸ showed that small-signal gain coefficients within a factor of two of those reported here could be produced with several joules of pump energy and, in addition, that the XeIII 109 nm gain can be

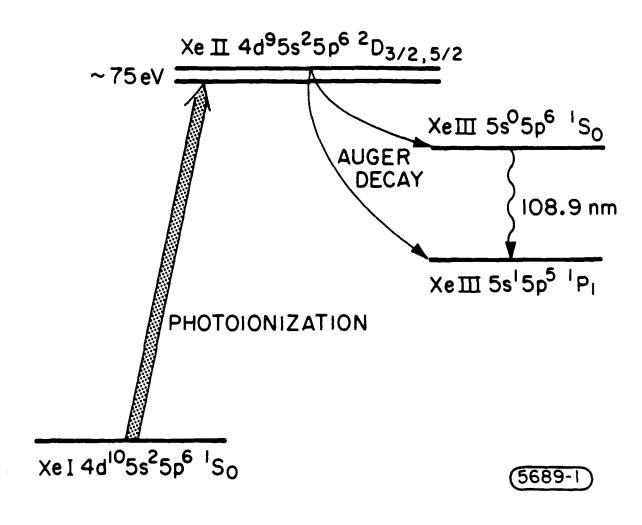


Fig. 1-Energy level diagram of Xe showing the levels relevent to photoionization and to Auger pumping of Xe III.

limited by competing processes. Their work suggests the most efficient use of pump energy requires a long, high aspect ratio geometry.

Figure 2 is a schematic diagram of the traveling-wave laser-produced-plasma excitation source. A 1064 nm laser is incident upon a cylindrical lens at $\theta = 68$ deg from normal and is focused onto a target which is parallel to the lens. This oblique focusing geometry has several advantages over the normal incidence arrangements used in previous work. ^{1.8} The large angle of incidence expands the length of the line focus by $1/\cos\theta$; therefore, our 3.3 cm diameter beam produces a 9 cm long plasma. In addition, the pump laser sweeps across the target, and the leading edge of the plasma travels at a speed, $c/\sin\theta$, only 8% greater than that of light. The emitted soft x-rays thus provide nearly synchronous traveling-wave excitation of the ambient gaseous medium.

In order to reduce the pump energy lost to grazing incidence reflection, grooves were cut into the target surface at a 45 deg angle, as shown in the inset of Fig. 2. The grooved surface decreases the local angle of incidence of the p-polarized pump laser from 68 deg to 23 deg and divides the input beam to form many small, separated plasmas rather than one continuous line. The combined length of these plasmas is only slightly greater than the input beam diameter. As a result, the extended gain length can be pumped with increased 1064 nm intensity and improved soft x-ray conversion efficiency.

All of the experiments described here were performed with a 3.5 J, 300 psec FWHM pump laser with a repetition rate of 1 shot every 5 minutes. The 3.3 cm diameter, spatially uniform, input beam was compressed (using normal incidence cylindrical optics) to 1.7 cm in the focusing dimension to increase the f-number of the lens and reduce aberrations. The focal length of the oblique cylindrical lens can be approximated by the sagital focal length of a tilted spherical lens; for $f_0 = 20 \, \text{cm}$ and $\theta = 68 \, \text{deg}$, the focal length is 12 cm. A 2.5 cm diameter stainless steel rod, threaded at 19 grooves cm⁻¹ and electroplated with gold, served as the target. This arrangement produced a focal line width of 200 μ m and an intensity on target of about $2 \times 10^{11} \, \text{W cm}^{-2}$. The ambient Xe pressure was 4 torr.

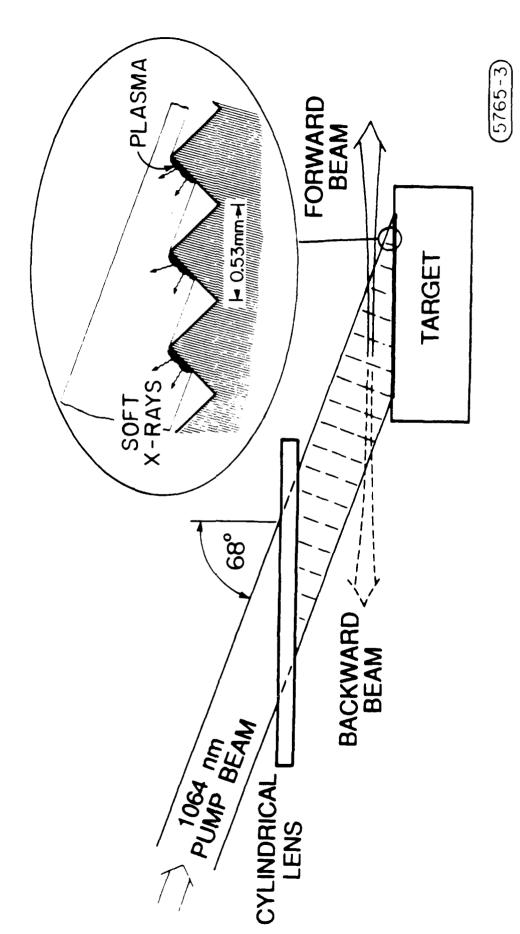


Fig. 2-Traveling-wave laser-produced-plasma soft x-ray source.

The observed excited volume was defined by two plates separated by 1.5 mm, through which the 1064 nm pump laser was focused, and by two 2 mm diameter pinholes on an axis 1.5 mm above the target and located 2 cm from the ends of the line focus. We monitored the 109 nm emission in the forward and backward directions simultaneously using two 0.2 m VUV monochrometers coupled to windowless channel electron multipliers. A 1 mm thick LiF window isolated each of the monochrometers from the Xe cell. To avoid saturation of the electron multipliers, we used calibrated LiF and O₂ gas cell attenuators to achieve the 10⁵ dynamic range required in these experiments.

The small-signal gain on the 109 nm transition was determined from measurements of time-integrated emission (the 109 nm pulses were shorter than the 700 psec response time of the detection system) as a function of length. The length of plasma on the target, and hence of the gain medium, was varied by masking the input laser beam. Figure 3 shows the increase in forward-propagating emission with length for three short sections of the target. A simple exponential fit to the data yields an average, time-integrated, small-signal gain coefficient of 4.4 cm⁻¹. This is a 70% improvement over the value obtained with a smooth, gold-plated target. Based on the measured, uniform small-signal gain coefficient, unsaturated amplification along the full 9 cm of length would provide a total gain of exp(40), or 170 dB.

The large-signal behavior of both the forward and backward 109 nm laser emission is shown in the semi-log and linear plots of Figs. 4a and 4b, where each symbol represents the average of at least three data points. For short gain lengths, the slopes of the forward and backward energy vs. length curves (on the log scale in Fig. 4a) are approximately the same. Beyond 4cm of length, the forward beam grows linearly (Fig. 4b) while the backward emission remains constant. This behavior indicates that the forward beam is fully saturated and is extracting nearly all the stored energy from the second half of the length.

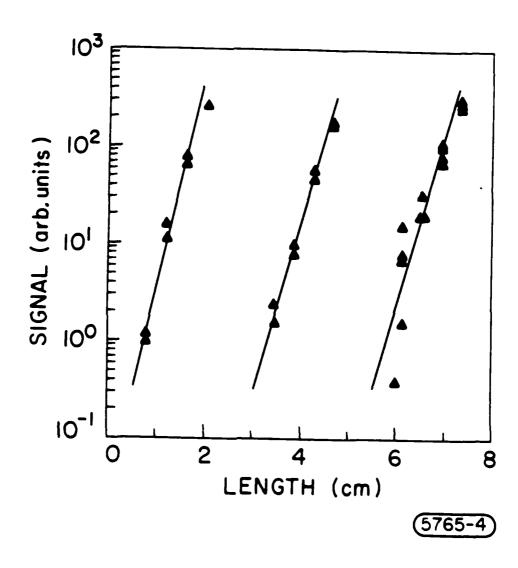


Fig. 3-109 nm signal versus length for three different sections of the target. The average exponential gain coefficient is $4.4 \, \mathrm{cm}^{-1}$.

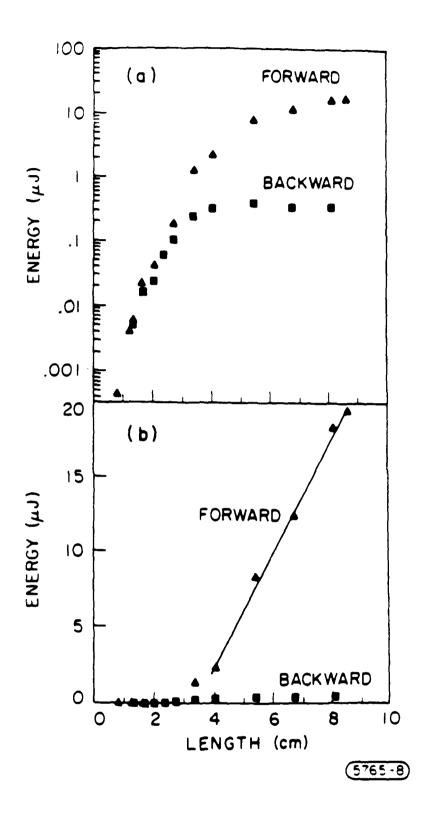


Fig. 4-109 nm energy versus plasma length on (a) log scale, (b) linear scale showing saturated, linear growth of the forward beam.

The vertical scale of Fig. 4 was calibrated in units of energy by replacing the monochrometer-based detection systems with a fast (350 psec), NBS-calibrated vacuum photodiode (Al₂O₃ photocathode) and calibrated LiF window. The increase of energy with length was identical to that in the emission measurements made using the monochrometers. The maximum energy output was $20 \,\mu$ J in the forward direction and $0.4 \,\mu$ J in the backward direction, yielding a forward-to-backward emission ratio of 50:1.

By visual observation of fluorescence on a scintillator located 90 cm from the target, and by translation of the vacuum photodiode in this plane, we estimate a forward beam divergence of 10 mrad. This small divergence is consistent with the large aspect ratio (length / width ≈ 60) of the geometry. The pulsewidth of the 109 nm laser emission was less than the 350 psec time resolution of the photodiode, which implies an output power greater than $50\,\mathrm{kW}$.

Assuming the measured energy is extracted predominantly from the last 6 cm of gain length, the total energy stored in the observed volume is $30\,\mu\text{J}$, or about 10^{-5} of the 1064 nm pump energy. Taking the cross sectional area of the laser to be $0.03\,\text{cm}^2$, we calculate an energy density of $110\,\mu\text{J}\,\text{cm}^{-3}$ stored in the 109 nm inversion. Given the atomic parameters of the system, i.e. an average 4d photoionization cross section of 15 Mb between 70 and 130 eV, the 5% Auger yield, and \sim 12% quantum efficiency, we can deduce a conversion efficiency of 1064 nm light to useful soft x-rays of approximately 2%.

The relationship of the observed gain behavior to the measured stored energy is complicated by the transient nature of the population inversion. The spontaneous lifetime of the upper level is 4.75 nsec,² but the inversion lifetime and pulse length are governed by stimulated decay and are on scale with the transit time of the gain medium. The large forward-to-backward emission ratio imparted by the traveling-wave excitation can be explained in terms of competition between the two beams. Although the slopes of the forward and backward energy vs. length curves in Fig. 4a are similar for the shorter lengths,

the forward beam reaches saturation earlier and, therefore, dominates in the second half of the length.

In this work we have demonstrated single-pass gain saturation of a photoionization-pumped laser. We have employed a traveling-wave laser-produced-plasma geometry which efficiently excites an extended gain length using only a few joules of pump energy. These results represent a significant step in the development of practical photoionization-pumped lasers.

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Figure Captions

- 1. Energy level diagram of Xe showing the levels relevent to photoionization and to Auger pumping of Xe III.
- 2. Traveling-wave laser-produced-plasma soft x-ray source.
- 3. 109 nm signal versus length for three different sections of the target. The average exponential gain coefficient is $4.4\,\mathrm{cm}^{-1}$.
- 4. 109 nm energy versus plasma length on (a) log scale, (b) linear scale showing saturated, linear growth of the forward beam.

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